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ON MACROSCOPIC DESCRIPTION OF PHOTONUCLEAR ISOTOPE PRODUCTION

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Based on an analytical model, a set of macroscopic parameters, that determine the activity of an isotopic target induced with a bremsstrahlung (X-ray) beam, is established. Those parameters can be measured experimentally with good accuracy. The proposed approach enables prompt estimate of the optimal dimensions and activation regime for the target of a given mass located at a given distance from a bremsstrahlung converter to ensure the maximum yield of an isotope product. As an example, the conditions for producing ^{99}Mo by bremsstrahlung with end-point energy of 40 MeV in a target from natural molybdenum by mass of 10…100 g positioned at a distance of 0…10 mm from the converter are examined. The analytical predictions are in good agreement with Monte Carlo simulations.

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INTRODUCTION

Production of a number of isotopes using a beam of high-energy bremsstrahlung (X-ray) photons is considered as a possible soft alternative to the traditional reactor-based technologies [1]. Such a method includes preliminary transformation of an electron beam with energy $E_e > E_{th}$, where E_{th} is the threshold of a reaction used, into the beam of bremsstrahlung photons. The photon energy E is continuously distributed over the range $0 < E \le E_e$. The X-rays act on an isotopic target followed by the A(γ,x)B reactions, where x can be n, p, np, α, etc.

Bremsstrahlung radiation can be generated either in a separate target-converter made of a high-Z material (like tantalum, tungsten), or in the isotopic target itself. The preliminary studies have shown that the second approach gives an advantage in the yield of the isotope product in the case of a thick target consisting of medium or heavy nuclei [2]. However, such an approach is not very effective when the target is made from a low-Z material or it is based on a metal oxide. In addition, the direct acting on the target with a high-current electron beam poses the problem of the target thermal stability.

In view of the weak attenuation of the high-energy gammas in the substance, and also X-rays angular divergence, the photonuclear reactions can take place practically within the semi-space. The corresponding distribution of the specific activity is essentially non-uniform with maximum on the radiation axis and tends to zero at the target periphery [3]. Therefore, the necessary stage of development of a photonuclear technology is optimization of the target geometry and conditions of target activation to ensure the maximum yield of the isotope product.

Commonly, the study of conditions for the photonuclear isotope production by bremsstrahlung radiation is carried out by Monte Carlo technique based on description of the elementary electron and photon interactions with matter [1]. Such an approach provides the data on the generated target activity and its distribution in the target for a specified set of input microscopic parameters. At a given statistical uncertainty, the accuracy of an end result is determined mainly by the precision of description of the reaction microscopic cross section.

The objective of this work is to establish a set of controllable macroscopic parameters that determine the total value and volumetric distribution of the target activity induced with a bremsstrahlung beam, to optimize the target activation conditions.

1. METHODS AND MATERIALS

1.1. MICROSCOPIC DESCRIPTION OF PHOTONUCLEAR ISOTOPE GENERATION

As it was preliminary mentioned, the process of isotope production by bremsstrahlung radiation can be described at a microscopic level. So, at the initial stage, an accelerated electron interacts with the electric field of a nucleus and orbital electrons of a converter followed by generation of a bremsstrahlung photon. In the case of a thick converter, the events of such type can occur several times until the electron to become thermalized or leaves the converter. In turn, the generated photon can either to leave the converter at once or preliminary interact with it changing the energy and momentum. Such an interaction can include the coherent (Rayleigh) and incoherent (Compton) scattering, photoelectric effect, generation of electron-positron pairs, and also photonuclear reactions [4]. The combination of such events at a given spatial and energy distribution of the primary electron beam, as well as the material and design of the converter determines the composition, angular and spectral distribution of the radiation leaving the converter and incident on the isotopic target.

For calculating the photonuclear yield by Monte Carlo technique, the trajectory of a photon with initial energy E in the target is represented as a series of the straight segments between the points of the photon interaction with the atoms. The length l_i of each i-segment (step) of the trajectory changes randomly near the photon free range $r_0(E)$, or

$$l_i \approx r_0(E) = (\mu(E)\rho)^{-1}, \qquad (1)$$

where $\mu(E)$ – is the mass attenuation coefficient for the photons with energy E determined by the total cross-section of the photon interaction with a target material, ρ – is the density of the material.

To determine the isotope yield, at the end of each trajectory segment l_i the probability of a single reaction, i.e. the event A(γ,x)B, is calculated by comparing the cross section of this reaction with the cross sections of all other electromagnetic interactions of the photon. It should be noted, that the contribution of the photonuclear processes to the total cross section of the photon interaction with matter does not exceed usually of several percent [4]. Therefore, for a given statistical uncertainty, the calculating time of the photonuclear yield depends on the microscopic cross section of the reaction and it is significantly longer than that of the spatial-energy distribution of the photon flux.

1.2. SEMI-MACROSCOPIC APPROACH TO CALCULATING TARGET ACTIVATION

Taking into account the last circumstance, a method based on a semi-macroscoping description of the photonuclear process was proposed to speed up the calculation of the photonuclear yield [5]. It was assumed that the photon energy E_i along each i-step of the photon trajectory in a target can be considered constant. In this case, the total isotope yield Y in the target can be represented as the sum of the micro-yields of the reaction along all the steps that make up the trajectory of each above-threshold photon in the target, namely,

$$Y = \sum_i \{1 - \exp[-\Sigma_x(E_i) \cdot l_i]\}, \quad (2)$$

where $\Sigma_x(E_i)$ – is the macroscopic cross section of the reaction A(γ,x)B, determined as

$$\Sigma_x(E_i) = \frac{N_A}{\overline{A}} \rho \nu \sigma(E_i), \quad (3)$$

N_A – is the Avogadro's number; $\overline{A}$ – is the average atomic weight of the target material; ν – is the relative content of an isotope-target; $\sigma(E_i)$ – is the microscopic cross section of the reaction A(γ,x)B. This approach enables to reduce the calculating time by approximately 4 orders of magnitude without reducing the accuracy of an end result.

The threshold of a single-nucleon photonuclear reaction is about 10 MeV [6]. The free range of the photons with such energy is about 1 cm or still more [7]. This value determines the scale of the isotope generation region and, consequently, the thickness of a technological target. Other basic parameters of a photonuclear technology such as the width of the electron and X-ray beams, the converter thickness, the distance from the converter to the target are also macroscopic and can be measured experimentally. Thus, transition from the micro- to macro-description of a photonuclear process enables to pass from Monte Carlo summations of a large number of individual particle interactions with matter, to the prompt analytical estimate of the yield of a photonuclear reaction based on a limited set of macroscopic parameters of the process. In particular, such an approach makes it possible to establish explicitly the dependence of the technology's capacity on those parameters and to optimize them.

1.3. MACROSCOPIC ANALYSIS OF TARGET ACTIVATION

It was shown in an analytical model (S-model-[3]), that for a case of practical interest of a cylindrical photonuclear target by radius R, the distribution of the linear activity of an isotope with decay constant λ, generated by a Gaussian electron beam with average current I for the irradiation period t can be calculated by expression

$$\frac{dA}{dz} = Y_0(E_e)\frac{I\rho}{e}[1-\exp(-\lambda \cdot t)] \times \left\{1 - \exp\left[-\frac{R^2}{2\delta_\gamma^2(z)}\right]\right\} \exp[-\mu(E_{\max})\rho(z-a)], \quad (4)$$

where

$$Y_0(E_e) = \frac{N_A}{\overline{A}} \nu \int_{E_{th}}^{E_e} n_\gamma(E_\gamma, E_e)\sigma(E)dE, \quad (5)$$

$$\delta_\gamma(z) = \delta_{\gamma,0} + z \cdot tg\theta_m, \quad (6)$$

E_{max} – is the photon energy corresponding to maximum of giante dipole resonance (GDR); $n_\gamma(E)$ – is the integrated-over-angle spectral distribution of the photons reduced to one primary electron; $\delta_{\gamma,0}$ – is SD of the flux density distribution of the above-threshold photons at the rear surface of the converter; Θ_m – is the photon most probable exit angle of such photons.

The factor Y_0 was called a coefficient of photonuclear conversion. In physical meaning, the Y_0 quantity corresponds to the reaction yield in a thin target normal to the X-ray beam and completely overlapping it, normalized to the one primary electron and mass thickness of the target. Such a target was called a photonuclear converter, PNC The Y_0 value can be calculated analytically (see Ref. [3]), determined by Monte Carlo simulation, and also measured experimentally with high accuracy using expression

$$Y_0^{\exp}(E_e) = \frac{eA_{PNC}(E_e,t)}{I \cdot d_{PNC}^m[1-\exp(-\lambda \cdot t)]}, \quad (7)$$

where A_{PNC} is the PNC activity, d_{PNC}^m is the PNC mass thickness.

From Eq. (4) one can derive the expression for the activity of a target by radius R and height H, located at a distance a from the bremsstrahlung converter,

$$A = Y_0(E_e)\frac{I\rho}{e}\left[1-\exp\left(-\lambda \cdot t\right)\right] \times \int_a^{a+H} dz \left[1 - \exp\left(-\frac{R^2}{2\delta_\gamma^2(z)}\right)\right] \exp[-\mu(E_{\max})\rho(z-a)]. \quad (8)$$

In the case of a target with given mass M, the connection is imposed on its height and radius. From here, one can obtain the equation for determining the optimal radius ensuring maximum target activity

$$\frac{\partial}{\partial R}\int_{a}^{a+\frac{M}{\pi\cdot\rho\cdot R^2}} dz\left\{1-\exp\left[-\frac{R^2}{2\left(\delta_{\gamma,0}+z\cdot tg\theta_m\right)}\right]\right\}\times$$
$$\times\exp[-\mu(E_{\max})\rho(z-a)]=0. \quad (9)$$

To make it possible the analytical calculation of the Eqs. (8), (9), we have replaced the integrand in them with its approximation, namely,

$$\left[1-\exp\left(-\frac{R^2}{2\delta_\gamma^2(z)}\right)\right]\exp[-\mu(E_{\max})\rho(z-a)]\cong$$
$$\cong\left[1-\exp\left(-\frac{R^2}{2\delta_\gamma^2(a)}\right)\right]\exp[-\mu_A(z-a)], \quad (10)$$

where

$$\mu_A\equiv\mu(E_{\max})\rho+\frac{\mathrm{tg}\theta_{\mathrm{m}}}{\delta_\gamma(a)}, \quad (11)$$

$\delta_\gamma(a)$ – is SD of the flux density of the above-threshold photons in the front plane of the target. It can be determined using expression

$$\delta_\gamma(a)=\left[\delta_e^2+\delta_{\gamma,\min}^2(a)\right]^{1/2}, \quad (12)$$

$$\delta_{\gamma,\min}(a)=\delta_{\gamma,0}^{\min}+a\cdot tg\theta_m. \quad (13)$$

Taking into account Eq. (10), the expression for the target activity takes the form

$$A(R,H)=\frac{dA}{dz}|_{z=a}\cdot\frac{1-\exp\left[-\left(\mu(E_{\max})\rho+\frac{tg\theta_m}{\delta_\gamma(a)}\right)\cdot H\right]}{\mu(E_{\max})\rho+\frac{tg\theta_m}{\delta_\gamma(a)}}, \quad (14)$$

where $\frac{dA}{dz}|_{z=a}$ – is the linear activity of the target's front part. It can be determined experimentally, and also calculated by Monte Carlo technique or analytically using expression

$$\left.\frac{dA}{dz}\right|_{z=a}=Y_0\frac{I\rho}{e}[1-\exp(-\lambda t)]\cdot\left[1-\exp\left(-\frac{R^2}{2\delta_\gamma^2(a)}\right)\right]. \quad (15)$$

It follows from Eqs. (14),(15), that the optimal radius of a target by mass M can be determined from the condition

$$\left.\frac{\partial\xi}{\partial R}\right|_{R=R_{opt}}=0, \quad (16)$$

where

$$\xi=\left[1-\exp\left(-\frac{R^2}{2\delta_\gamma^2(a)}\right)\right]\cdot\left[1-\exp\left(-\frac{\mu_A M}{\pi R^2\rho}\right)\right]. \quad (17)$$

In a physical sense, the quantity ξ corresponds to the enclosed in a (M,R)-cylinder the part of a theoretical limit of the activity generated in a conventional half-space target

It is evident from Eqs. (14)-(17), that the activity of the target, in addition to its dimensions and distance to the converter, depends also on the width of the above-threshold photon beam incident on the target. The optimal photon beam width can be determined from the equation

$$\frac{\partial\xi}{\partial\delta_\gamma(a)}=0. \quad (18)$$

2. RESULTS AND DISCUSSION

2.1. VALIDATION OF METHODS FOR DETERMINING THE TOTAL ACTIVITY AND ITS DISTRIBUTION IN THE TARGET

The accuracy of the analytical model predictions was evaluated by comparing them with experimental results and Monte Carlo simulations [8]. The different converter and target options were considered. To determine the activity linear distribution with target depth (z-a), a design of the accelerator exit device presented in Fig.1 was developed. Behind an electron accelerator exit window 1 (Ti, 50 μm), an aluminum pipe 400 mm long and 60 mm in diameter coaxial with the electron beam was positioned. Inside the pipe the converter 2 and target 3, fastened with the aluminum elements, was accommodated. The converter comprised the four tantalum plates each 1mm thick, separated by the same air gaps for cooling. The target was performed as a cylindrical stack from natural molybdenum by 20 mm in diameter and 23.8 mm in height, consisting of alternating foils and disks by 0.1 and 2.9 mm in thickness, respectively. The foils were used to measure the induced ^{99}Mo activity by γ-spectrometry technique with minimum self-absorption.

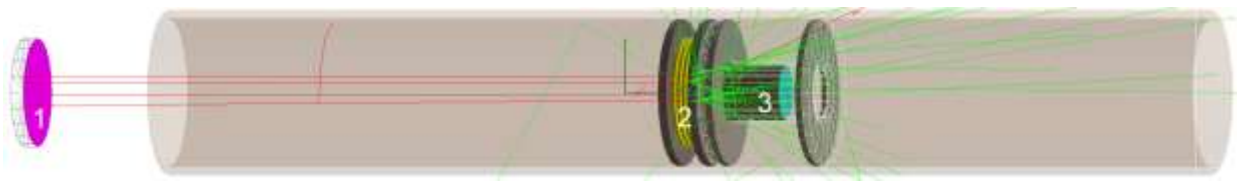


Fig. 1. Geometry of the accelerator exit device used in the experiment and simulations

The conditions of the target activation and measurement of foil activity are described elsewhere [9]. Fig. 2 shows the activity distribution along the target axis, obtained by Monte Carlo method, and also using the Eqs. (8) and (14) of the model. It is evident, that the original expression (8) describes the activity profile somewhat better than its simplified version (14). At the same time, both variants give some overestimation in the activity of the target rear part. This can be explained by the escape of some above-threshold

photons through the side surface of the target due to the Compton scattering. Considering that the main mechanism of the interaction of photons with energy of about 10 MeV and higher with the substance is generation of electron-positron pairs, the proposed model does not take into account the scattering of such photons. At the same time, the rapid decrease of linear activity along the target axes makes the contribution of the target rear part to the total target activity insignificant. In particular, the measured total target activity is 554.1 μCi, the value obtained by Monte Carlo simulations is 540.0 μCi, while the model calculations using Eqs. (8) and (14) give 553.2 and 565.7 μCi, respectively. Thus, the difference in the results obtained by different methods does not exceed 5%.

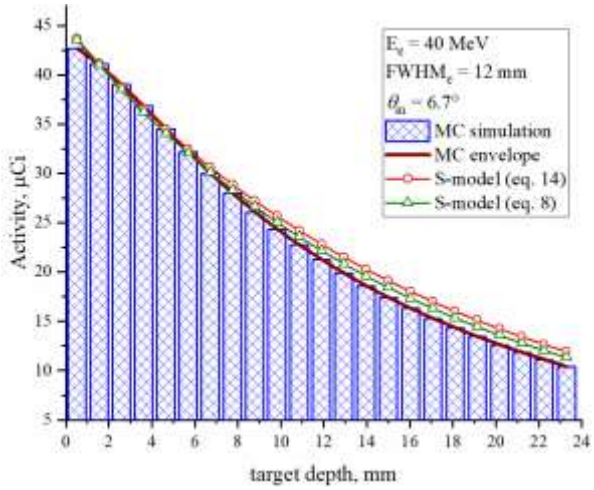


Fig. 2. Distribution of ^{99}Mo linear activity in the target-assembly from natural molybdenum

2.2. OPTIMIZATION OF TARGET GEOMETRY

As an example of optimization of the target geometry, the results of the calculation of optimal radius for a 10 g target from natural molybdenum using Eqs. (16), (17) are presented Fig. 3. The target is positioned at a distance of 10mm from a solid tungsten converter by 3.5 mm in thickness. The converter is exposed to an electron beam with energy 40 MeV and width 12 mm (FWHM). It is evident that the optimal radius of the target is about 6 mm.

Table 1 lists the data on the optimal radius R_{opt} and height H_{opt}, as well as the corresponding activity A for the targets of different mass from natural molybdenum obtained by the numerical solution of both the original Eq. (8), and also of its simplified representation by Eq. (14). For comparison, the data on the target activity A^{MC} obtained for the same conditions by Monte Carlo simulations [8] are given as well. It is evident, that the distinction in the optimal target radius obtained by different methods does not exceed 6 %, while the corresponding values of the target activity are differed not greater than by 3 %. At the same time, the absolute maximum of the activity predicted by the model is overestimated compared to simulations by up to 5 %.

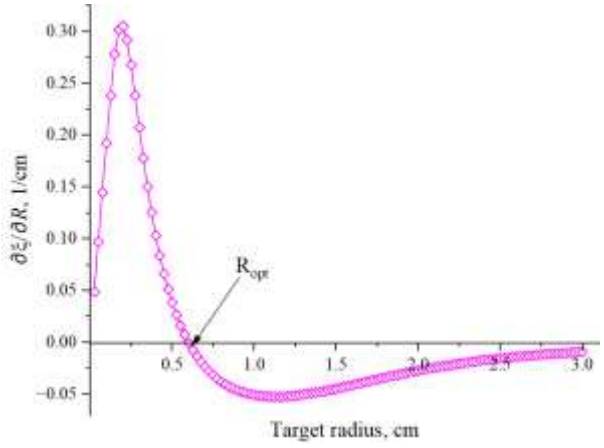


Fig. 3. Dependence of the derivative $\frac{\partial \xi}{\partial R}$ on target's radius

2.3. DEPENDENCE OF OPTIMAL TARGET SIZE ON WIDTH OF THE PHOTON BEAM

The results of calculating the reduced to the charge of the electron beam and averaged specific activity (A_{mass}, MBq/μA·h·g) of a target located close to a solid 3.5 mm thick tungsten converter depending on the target radius and SD of the photon flux density on target's input, $\delta_{\gamma,0}$, are shown in Fig. 4.

Table 1

Yield of ^{99}Mo in the targets of various mass and optimized dimensions

Target mass, g	Eq. (8)				Eq. (14)			
	R_{opt}, mm	H_{opt}, mm	A, kBq/μA·h	A^{MC}, kBq/μA·h	R_{opt}, mm	H_{opt}, mm	A, kBq/μA·h	A^{MC}, kBq/μA·h
10	6.93	6.49	2404.1	2386.1	6.54	7.28	2475.4	2385.4
20	8.24	9.17	4109.6	4042.4	7.78	10.29	4234.7	4054.2
50	10.36	14.51	7723.8	7465.5	9.78	16.28	7874.6	7475.0
100	12.32	20.52	11583.7	10995	11.63	23.03	11531.4	11060.8

The maximum local specific activity in the target obtained by simulations, Max MC, is shown, as well as the data calculated by Eqs. (16),(17) and denoted by "Opt" (the value of the corresponding average specific activity is given next in parentheses). It is evident, that the optimal target radius predicted by the model is close to that obtained by simulations. It should be noted also, that the dependence of the reaction yield on the target dimensions and width of the photon beam is more pronounced for the light targets.

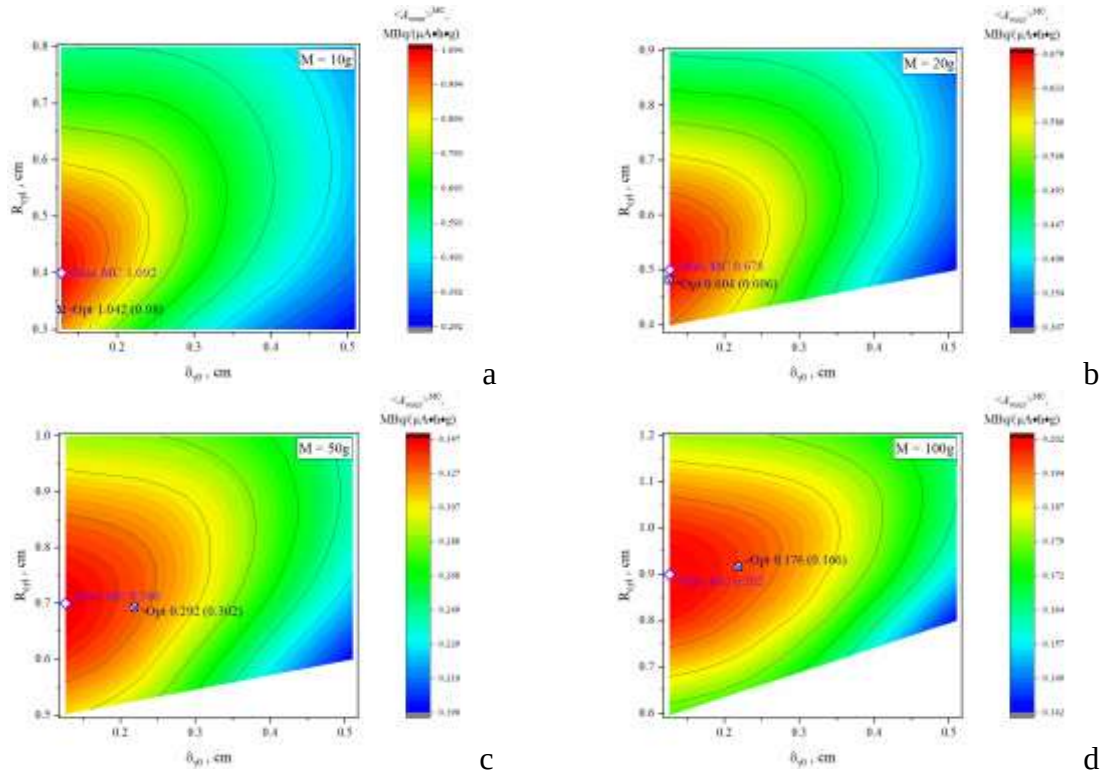


Fig. 4. Average specific activity of ^{99}Mo in natural molybdenum targets, placed close to the converter, depending on target radius and width of the above-threshold photon beam:
a – M = 10 g; b – M = 20 g; c – M = 50 g; d – M = 100 g

2.4. DEPENDENCE OF OPTIMAL TARGET GEOMETRY ON DISTANCE TO CONVERTER

Fig. 5 shows the results of calculation by the Eqs. (12) – (17) of maximum specific activity A_{max} and optimal radius R_{opt} of a target located at a distance a from the converter depending on SD of the photon flux density on the front surface of the target, $\delta_\gamma(a)$.

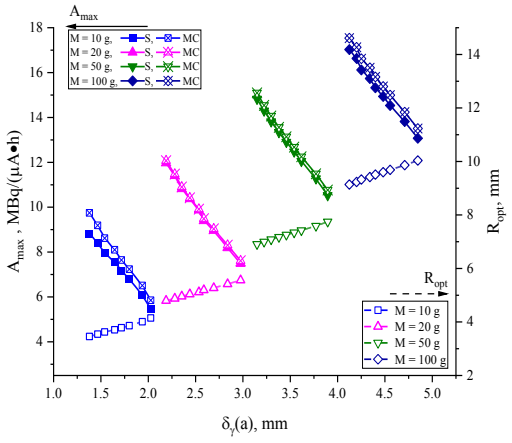


Fig. 5. Dependence of A_{max} and R_{opt} on SD of the photon flux density distribution on the target (filled and empty dots correspond to S-model, crossed dots – simulation)

As it can be seen from Eqs. (12),(13), the $\delta_\gamma(a)$ value, apart from the distance to the converter, depends on SD of the photon flux density at converter's output, $\delta_{\gamma,0}$, and also on the photon most probable exit angle, Θ_m. The last two parameters are determined by width and energy of the electron beam, as well as material and thickness of the converter [10]. It is evident, that with an increase in the target mass, the optimal width of the photon beam increases as well. At the same time, for a given mass, with the growth of the photon beam width the optimal target radius comes up, while the target activity drops. Comparison of the model predictions with Monte Carlo simulations shows that the greatest underestimation of the model is less than 10% for the light targets and narrow electron beam ($\delta_e \sim 1\ldots2$ mm). In such a case, the distribution of photon flux density is described by the linear combination of the Lorentzian and Gaussian [11]. The exact Gaussian distribution is realized at $\delta_e \geq 2$ mm, where the best agreement between the model predictions and simulations takes place.

Table 2 presents the results of the model calculation and Monte Carlo simulations of the optimal dimensions R_{opt}, H_{opt}, as well as corresponding surface area S_{total} and activity of the targets, positioned at different distance from the converter at an optimal width of a 40 MeV electron beam. The data on the absorbed radiation power in the target P_{dep} and power flux density through the target front surface P_{in} are presented as well. Fig. 6 shows the results of calculating the average specific activity of the targets of different mass at the optimal target radius and SD of the photon flux density, predicted by the model, and also obtained by simulations. It is evident that the predicted region of the maximum reaction yield is satisfactory agreed with simulations.

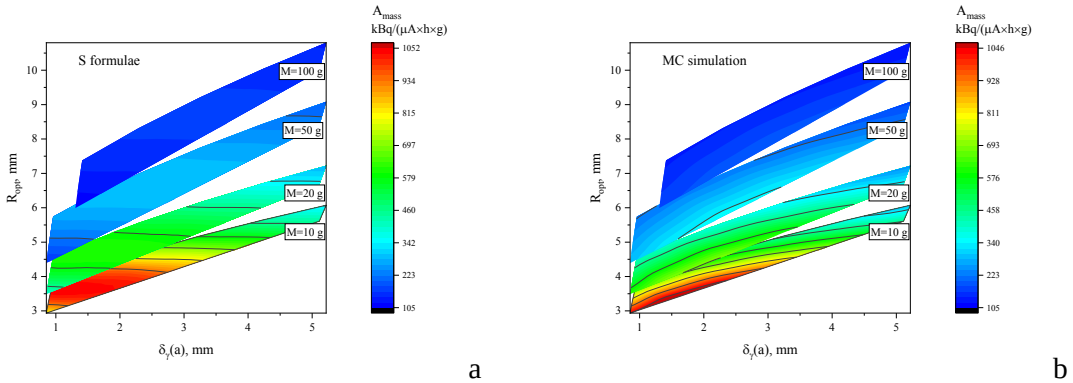


Fig. 6. Average specific activity of ^{99}Mo in the targets of different mass and optimal radius R_{opt}, at optimal photon beam size δ_γ (a), obtained analytically (a) and by simulations (b)

Table 2

Optimal conditions for producing ^{99}Mo in the cylindrical targets from natural Mo

a, mm	R_{opt}, mm	H_{opt}, mm	S_{total}, cm^2	P_{dep},, W/μA	P_{in}, W/cm^2·μA	$A^S_{\max}$, MBq/μA·h	$\langle A_{mass}\rangle^S_{\max}$, MBq/μA·hg·	A^{MC}, MBq/μA·h	$\langle A_{mass}\rangle^{MC}$, MBq/μA·hg·
M=10 g, δ_e, = 1 mm									
0	3.35	27.7	6.54	6.96	1.06	9.81	0.981	10.42	1.04
5	3.79	21.7	6.07	4.08	0.67	7.15	0.715	7.64	0.76
10	4.15	18.1	5.80	2.82	0.49	5.56	0.556	5.85	0.58
M=20 g, δ_e, = 2 mm									
0	4.81	26.8	9.58	8.35	0.87	12.1	0.606	12.09	0.60
5	5.20	23.0	9.23	5.63	0.61	9.19	0.460	9.49	0.47
10	5.56	20.1	8.98	4.09	0.46	7.33	0.367	7.62	0.38
M=50 g, δ_e, = 3 mm									
0	6.93	32.4	17.1	10.48	0.61	14.6	0.292	15.12	0.30
5	7.34	28.8	16.7	8.04	0.48	12.2	0.244	12.68	0.25
10	7.75	25.9	16.4	6.28	0.38	10.3	0.205	10.76	0.21
M=100 g, δ_e, = 4 mm									
0	9.16	37.1	26.6	12.13	0.46	16.6	0.166	17.61	0.17
5	9.60	33.8	26.1	10.02	0.38	14.7	0.147	15.38	0.15
10	10.04	30.9	25.8	8.29	0.32	12.8	0.128	13.51	0.13

CONCLUSIONS

The obtained analytical expressions for the total activity of a cylindrical molybdenum target induced by X-rays, as well as for linear activity distribution contain in a factorized form their dependence on nuclear physical parameters Y_0, σ, λ, E_{max}, ν, radiation physical parameters μ, δ_γ, θ_m, mechanical characteristics ρ, M, R, H, a, as well as on the characteristics of the target activation mode I, E_e, δ_e, t. All these quantities can be divided into those controllable, like Y_0, δ_γ, v, θ_m, I, E_e, t, a, M, R, H, and referenced, like σ, λ, E_{th}, E_{max}, μ. In turn, the controllable parameters comprise those specified directly I, E_e, δ_e, v, a, M, R, H, and also their derivatives Y_0, δ_γ,, θ_m.

The data on the microscopic cross section of a photonuclear reaction $\sigma(E)$ are the most problematic, as they differ sometimes from 30 to 100% and still more [12-14]. The last circumstance imposes the limitation on the accuracy of determination of the isotope yield via the microscopic description of a photonuclear process. Conversely, the analysis using the set of key macroscopic parameters Y_0, δ_γ,, θ_m, which can be measured experimentally with good accuracy [11], enables prompt and accurate estimate of the target activity, as well as its optimal dimensions and bremsstrahlung beam width ensuring maximum yield of an isotopic product.

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ПРО МАКРОСКОПІЧНИЙ ОПИС ФОТОЯДЕРНОГО НАПРАЦЮВАННЯ ІЗОТОПІВ

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На основі аналітичної моделі встановлено набір макроскопічних параметрів, що визначають індуковану пучком гальмівного випромінювання активність ізотопної мішені та її лінійний розподіл. Ці параметри можна виміряти в експерименті досить просто і з хорошою точністю. Запропонований підхід дозволяє проводити попередню оцінку оптимальних розмірів та умов активації мішені наданої маси, що розміщена на наданій відстані від конвертера гальмівного випромінювання, для забезпечення максимуму виходу ізотопного продукту. Як приклад, вивчені умови напрацювання ^{99}Mo гальмівним випромінюванням з граничною енергією 40 МеВ у мішені з природного молібдену масою 10…100 г, що розміщена на відстані 0…10 мм від конвертера. Результати аналітичних розрахунків добре погоджуються з даними Монте-Карло-моделювання.